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METALLIC GLASSES

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RESISTIVITY MINIMA IN  $\text{Fe}_x\text{Ni}_{1-x}/_{75}\text{B}_{25}$  METALLIC GLASSES

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#### АННОТАЦИЯ

Проводились очень точные измерения электросопротивления двух аморфных сплавов  $\text{Fe}_x\text{Ni}_{1-x}/_{75}\text{B}_{25}$  различного состава в области температур 1,5-50 К. Все сплавы ферромагнитные  $/x > 0,25/$  и имеют минимум сопротивления ниже температуры 30 К. Более глубокие минимумы дает средняя часть области исследуемых составов. Результаты сравниваются с аналогичными данными для системы  $\text{Fe}_x\text{Ni}_{1-x}/_{80}\text{B}_{20}$  и изучается взаимосвязь экспериментальных данных с теоретическими моделями, описывающими минимум сопротивления.

#### KIVONAT

Nagy pontosságu elektromos ellenállásméréseket végeztünk két különböző összetételű  $(\text{Fe}_x\text{Ni}_{1-x})_{75}\text{B}_{25}$  amorf ötvözetben az 1,5-50 K hőmérséklettartományban. Valamennyi megvizsgált ötvözet ferromágneses  $(x > 0,25)$  és ellenállás-minimumot mutat 30 K alatt. A minimumok mélyebbek az összetétel tartomány középső részén. Eredményeinket összehasonlítjuk a  $(\text{Fe}_x\text{Ni}_{1-x})_{80}\text{B}_{20}$  rendszerben találtakkal és megvizsgáljuk a kísérleti eredmények kapcsolatát az ellenállás-minimum leírását célzó elméleti modellekkel.



## ABSTRACT

High precision resistivity measurements were performed on seven  $\text{Fe}_{1-x}\text{Ni}_x/\text{B}_{25}$  alloys in the temperature range 1.5-50K. All the alloys are ferromagnetic  $/x \geq 0.25/$  and exhibit the resistance minima below 30K. The resistance minima are deeper for the intermediate  $x$  values. These and other results are compared with those for  $\text{Fe}_{1-x}\text{Ni}_x/\text{B}_{20}$  alloys and discussed in terms of the theoretical models which attempt to explain the resistance minima in metallic glasses.

## INTRODUCTION

It is well known that in some metallic glasses there is an upturn in the resistance at the lowest temperatures, i.e. a resistance minimum occurs. Although this phenomenon has been investigated rather extensively, there is no agreement so far even as to which /structural or magnetic/ excitations are responsible for this behaviour. This is not surprising since our knowledge of the electron transport in amorphous metals is rather restricted and on the other hand the experimental facts regarding the resistance minima are controversial [1]. To illustrate this we note the following observations:

- 1./ Minima of this type are only observed in the metallic glasses which contain 3d elements /Mn, Cr, Fe, Co or Ni/ or rare earths.

- 2./ More pronounced minima occur in the alloys with the 3d elements which are left from Fe in the periodic table.

- 3./ A small addition of Cr or Mn to strong ferromagnets of the  $\text{Fe}_{80}\text{M}_{20}$  type /where M is a combination of metalloids/ strongly enhances the upturn in the resistivity.



4./ Similar minima are observed in some crystalline alloys as non-stoichiometric  $\text{Fe}_3\text{Si}$  and disordered PtCr alloys.

5./ The upturn in resistivity decreases and eventually disappears on crystallization of glassy alloys.

6./ These upturns are practically independent of the magnetic field in ferromagnetic alloys, show only slight field dependence in the paramagnetic samples but may depend rather strongly on magnetic field in the intermediate cases.

Here we present the systematic study of the low temperature resistivities of  $\text{Fe}_x\text{Ni}_{1-x}/_{75}\text{B}_{25}$  glassy alloys and compare with our results obtained on  $\text{Fe}_x\text{Ni}_{1-x}/_{80}\text{B}_{20}$  series [2].

#### EXPERIMENTAL AND RESULTS

The samples were melt spun ribbons of appr. 0.5 mm wide and up to 30  $\mu\text{m}$  thick. The absolute resistivity values /given in Table 1. together with other relevant data/ were determined by measuring density, mass and length of our samples. The resistivity values are accurate to 2%, the actual resistance values were measured with a resolution of two parts per million.

The relative changes in the resistivity,  $\frac{\Delta\rho}{\rho_m} = (\rho - \rho_m) / \rho_m$  /where  $\rho_m$  is the resistivity value at the minimum/ of our alloys are shown in Fig. 1. The resistance minima temperatures which range 12-30 K are shown in Table 1. The values are practically constant for  $0.5 < x < 0.8$  and decrease rapidly for  $x < 0.5$ . Below the minimum the resistivities vary approximately logarithmically with temperature, the slopes/ expressed in  $\mu\Omega \text{ cm}$  per decade of temperature/ exhibit a broad maximum around  $x=0.5$ . In the alloys with intermediate  $x$  values  $/0.25 < x < 0.8/$  the logarithmic slopes show a slight temperature dependence around 4K. Above the minimum resistivity increases rapidly with temperature at a rate which depends on  $x$ . The resistivity variations above the minimum are shown separately in Fig. 2.

It can be seen that the total change in resistivity,  $\Delta\rho = \rho - \rho_m$  of none of these alloys obeys a simple power law in this temperature range. On the other hand, it was often claimed that



Table 1. Data relevant to  $\text{Fe}_x\text{Ni}_{1-x}/_{75}\text{B}_{25}$  metallic glasses:  
 $\rho_{4.2}$  is the residual resistivity, A is the logarithmic slope,  $T_m$  is the temperature of the resistance minimum, B is the coefficient of the  $T^{3/2}$  resistivity variation above the minimum and  $T_C$  is the Curie temperature

x	$\rho_{4.2}/\mu\Omega\text{cm}/$	$A/\mu\Omega\text{cm}/\text{decade}/$	$T_m/\text{K}/$	$B/\text{n}\Omega\text{cm}/T^{3/2}/$	$T_C/\text{K}/$
0.20	120	0.128	11.5	2.5	284
0.33	122.5	0.206	20	1.75	413
0.5	124.5	0.211	29	1.1	611
0.67	125.5	0.207	29.5	0.9	710
0.75	126	0.206	29.5	0.9	§
0.8	126.2	0.193	30	0.83	§
1.0	126.5	0.145	20	0.72	730 <sup>&amp;</sup>

§: Curie point cannot be determined by calorimetry due to the proximity of crystallization

&: Data from: T. Kemény et. al., Phys. Rev. B 20, 476 /1979/



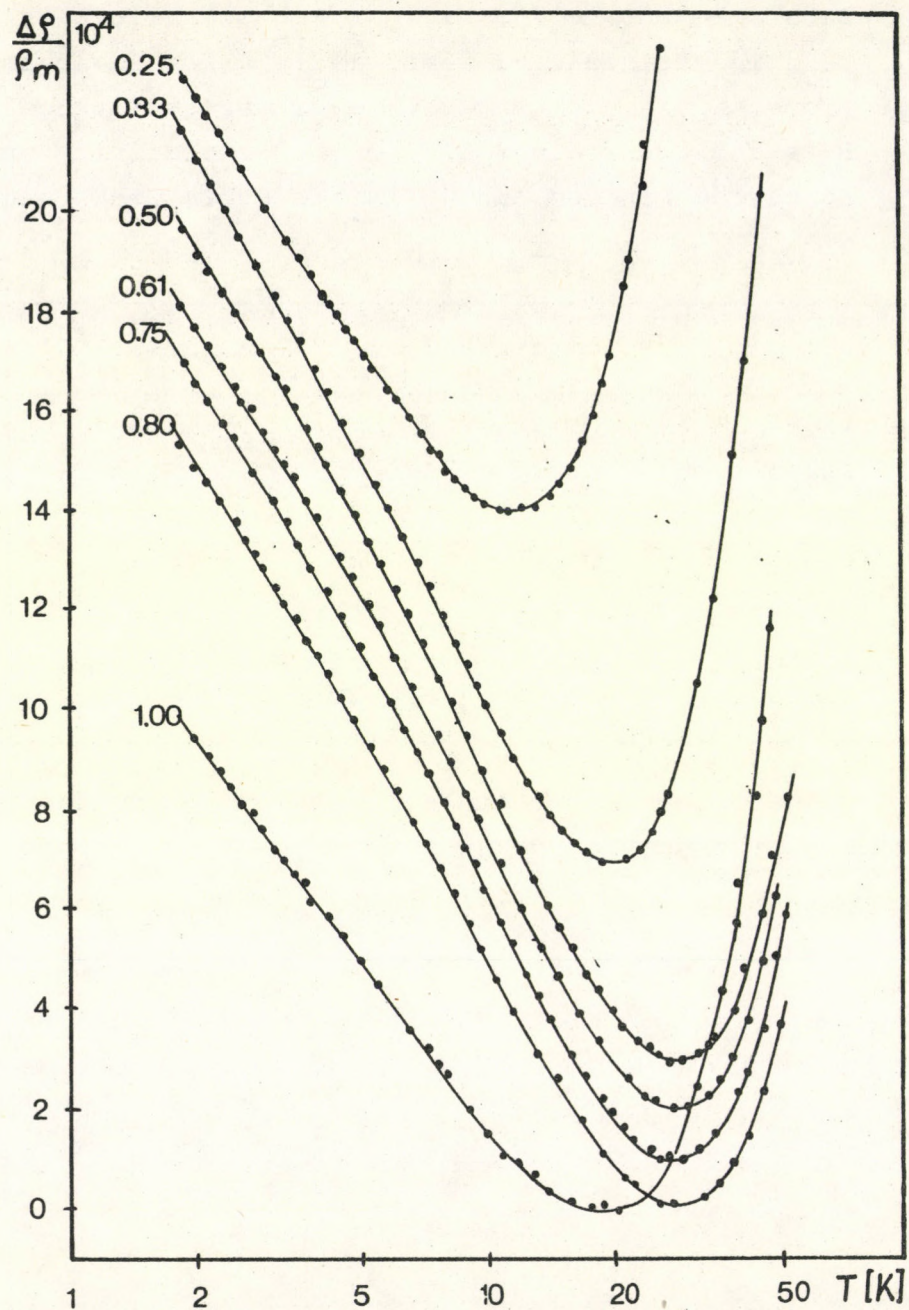


Fig. 1. Relative change in the resistivity of  $\text{Fe}_x\text{Ni}_{1-x}/75\text{B}_{25}$  glassy alloys. The numbers denote  $x$ .



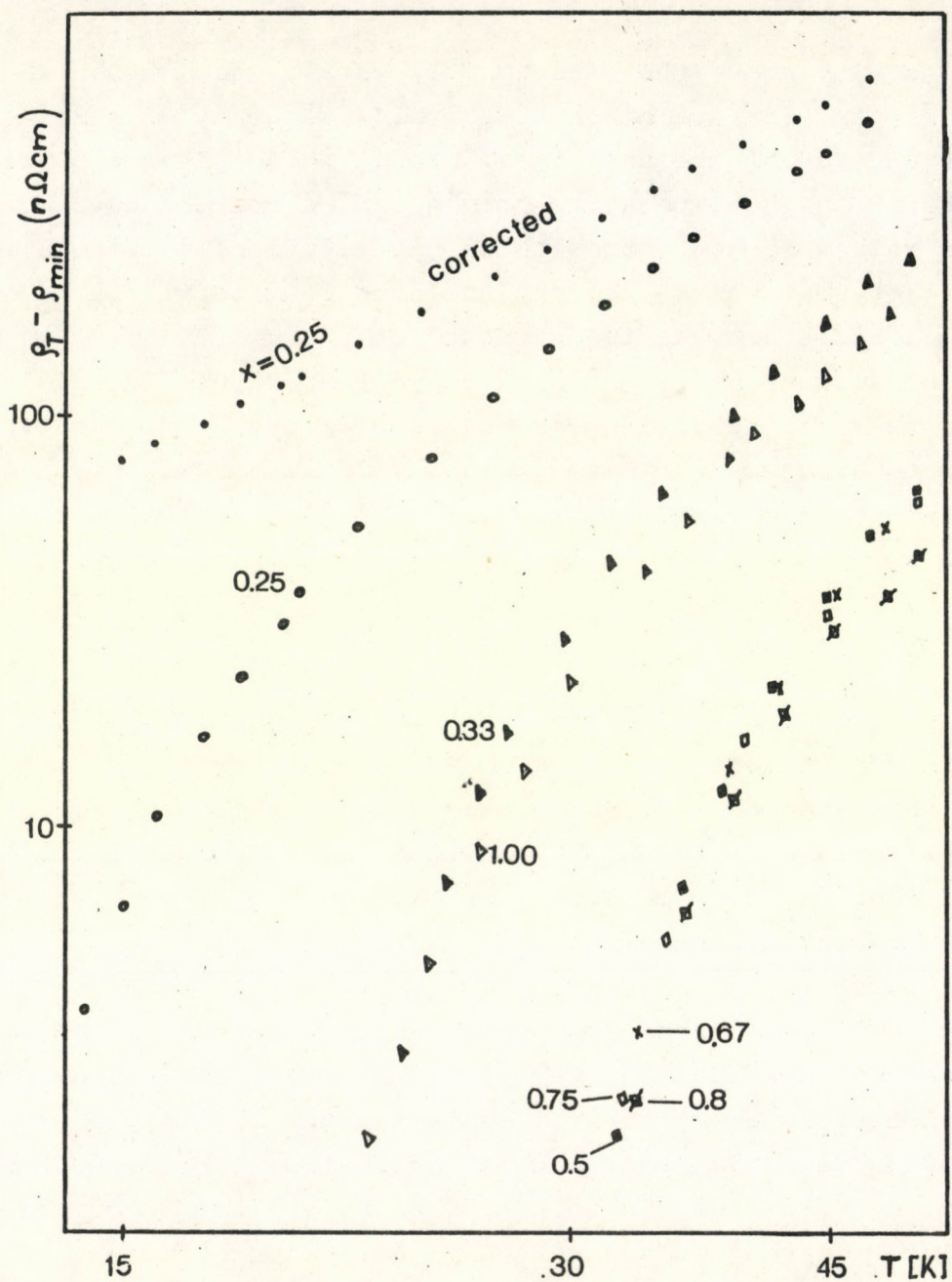


Fig. 2. The resistivity change of  $\text{Fe Ni}_{1-x} \text{B}_{25}$  glassy alloys above the minimum. The numbers denote  $x$ . The curve at the top shows the resistivity of  $x=0.25$  alloy corrected for the logarithmic contribution.



the resistivity above the minimum follows a  $T^2$  law, which we believe is an artifact of the rather low accuracy of some measurements and /or of the use of a  $T^2$  scale which gives too much weight to the high temperature points. If we correct however our measured values for the logarithmic contribution/ extrapolated from the low temperature region/ the data seem to fit much better to a simple power law as illustrated for the  $x=0.25$  alloy in *Fig. 2*. Our measurements in an extended temperature interval [2] indicate that the resistivities of these alloys vary roughly as  $T^{3/2}$  /up to about  $T_C/3$ , where  $T_C$  is the Curie temperature, see Table 1./ with the coefficient decreasing with  $x$  /Table 1./.

## DISCUSSION

As pointed out earlier the experimental facts about the resistance minimum in metallic glasses are somewhat controversial, some of them seem to favour magnetic while the other ones the structural origin of the anomalous resistivity behaviour. It must be noted that magnetic models often contain the structural disorder as an essential ingredient.

The structural models are based on the fact, that the amorphous state is not unique /in contrast to the crystalline one/ and therefore the tunneling may provide the fundamental excitation mechanism in amorphous solids [3,4]. One of the first and still most widely used calculations of the low temperature resistivity along this line [5] is based on the scattering of the conduction electrons on tunneling states. Although this calculation provides an expression which described qualitatively most of the experimental data the calculation itself may be criticised on several grounds [1]. Recent calculations of a more realistic version of this model without any internal-spin degree of freedom [6] seem to indicate that the resistivity contributions arising from this mechanism /taking experimental densities of tunneling states/ would be negligible. More recent calculations of the tunneling model take into account the fact that the operators describing the interaction between the conduction electrons and the tunneling systems do not commute in the momentum space [7].



A resistivity contribution is predicted which is proportional [7] to  $(\ln/T)^2$ , but as its magnitude depends on the coupling constants which are not yet known, no quantitative discussion of our results is possible along these lines.

The fact, that the minima appear only in alloys which contain a component with non-zero magnetic moment /3d or rare-earth element/, lead naturally to the use of Kondo model /for a review see [8]/ for their explanation. It is quite clear that the single impurity Kondo effect can explain rather well the resistance minima in PdSi amorphous alloys containing a small amount of 3d elements, see e.g. [9], but the model runs into difficulties in a strong ferromagnet as  $\text{Fe}_{80}\text{B}_{20}$ . The spin-flip scattering which is responsible for the Kondo effect must be quenched in the strong internal fields /100 T/ characteristic to ferromagnetic materials. It was argued however, that the structural randomness together with the superexchange mediated by the metalloid atoms produce a distribution of molecular fields where a small percentage of magnetic atoms may find themselves in vanishingly small magnetic field [10].

It is apparent that this model may qualitatively explain our results. Assuming that only Fe atoms carry a significant moment in our alloys one would expect that decreasing Fe content leads to more atomic sites with vanishing internal fields but at the same time there would be also less magnetic atoms to occupy those sites. One would therefore expect the strongest effect at some intermediate Fe content as it is observed. The comparison of the present results with those of  $\text{Fe}_x\text{Ni}_{1-x}/_{80}\text{B}_{20}$  indicates a tendency resistance minima are usually deeper in the alloys containing 25 at % B. This also in agreement with the model mentioned above, as it predicts that the probability of vanishingly small magnetic fields should increase with increasing metalloid content.

We have observed that the logarithmic slopes depend slightly on temperature around 4 K. The investigations are under way to decide whether it is connected with the transition between the different temperature regimes of the spin-flip scattering problem [8] or it indicates the appearance of a different scattering mechanism. The rather complicated temperature dependence of the



electrical resistivity in the low temperature region may easily indicate the presence of two /or more/ interactions. One of them may be a Kondo-like one as shown in  $\text{Fe}_x\text{Ni}_{80-x}\text{P}_{14}\text{B}_6$  alloys [11] by electrical resistance measurements in magnetic field while the exact origin of the other scattering mechanism is not yet known. The relative importance of different interactions is definitely dependent on composition. An almost single impurity-like Kondo effect is expected in the dilute limit,  $x \ll 1$  /somewhat complicated with the occurrence of some magnetic scattering also due to Ni clusters/. This effect is gradually eliminated by internal fields as the ferromagnetism appears, leaving only the contribution from that small percentage of magnetic moments which are in very small magnetic field due to the peculiar features of the distribution.

Finally we note that a strong  $x$  dependence of the resistivity variation above the minimum indicates a significant magnetic contribution to the resistivity also in this temperature range. The coefficient of the  $T^{3/2}$  term /Table 1./ shows a rapid increase with increasing  $x$ . Since the alloys with the lowest Fe content have also the lowest Curie temperature it is very probable that this term is mainly caused by electron-magnon scattering which is stronger in the alloys with lower  $T_C$  values.

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